

Island Density in Homoepitaxial Growth: Improved Monte Carlo Results

H. Jeong¹, B. Kahng², and D. E. Wolf³

¹ *Department of Physics and Center for Theoretical Physics,*

Seoul National University, Seoul 151-742, Korea

² *Department of Physics and Center for Advanced Materials and Devices,*

Kon-Kuk University, Seoul 143-701, Korea

³ *FB 10, Gerhard-Mercator Universität, D-47048 Duisburg, Germany*

Abstract

We reexamine the density of two dimensional islands in the submonolayer regime of a homoepitaxially growing surface using the coarse grained Monte Carlo simulation with random sequential updating rather than parallel updating. It turns out that the power law dependence of the density of islands on the deposition rate agrees much better with the theoretical prediction than previous data obtained by other methods if random sequential instead of parallel updating is used.

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Recently problems of surface growth by molecular beam epitaxy (MBE) have attracted a lot of interest [1–4]. From the point of view of statistical physics it is intriguing, how various stochastic processes such as shot noise, diffusion, and nucleation, give rise to scaling structures involving many atoms. These cooperative phenomena depend crucially on the ratio between the diffusion constant D and the deposition rate F (number of atoms landing on the surface per unit area and unit time). If adatoms can diffuse to stable positions such as kink sites or step edges, before other adatoms get deposited, and if interlayer diffusion is not inhibited by Ehrlich-Schwoebel barriers [5,6], smooth surfaces with a minimum of defects can be grown. High symmetry surfaces exhibit layer-by-layer growth called also Frank-van-der-Merve mode [7]. However, when the deposition rate is high compared to the hopping rate of adatoms, the surface gets rough quickly.

In the present note we reconsider the structures formed when less than a monolayer is deposited. It has long been known that the maximum density of islands, before coalescence starts, has a power law dependence $\rho \propto (D/F)^{-2\gamma}$ [8–11]. Using rate equations, the exponent γ can be predicted. If only adatoms can move and desorption can be neglected, one obtains [10,11]

$$\gamma = \frac{i^*}{2i^* + d + d_f}, \quad (1)$$

where $s^* = i^* + 1$ is the size of the smallest stable island, which means that an island with more than i^* atoms is more likely to grow than to shrink. d is the surface dimension and d_f the fractal dimension of the islands [12].

This prediction has been supported by many computer simulations for various models [13–17]. In particular for large i^* the exponents were found to be slightly (up to 10%) smaller than the theoretical prediction. The question has been raised, whether this must be so, because islands smaller than s^* may also be stable with a rather high probability, so that the exponent might be determined by an effective $i_{\text{eff}}^* < i^*$. Here we present new data which indicate that with a slight modification of the simulation model used in [17] the γ -values turn out to be much closer to the theoretical prediction than before.

Numerical simulations of MBE require large computation times, because the deposition of one monolayer takes very long on the time scale of the adatom diffusion. Moreover, it is easy to see that the system size required in order to avoid strong finite size effects, must be larger than $\max(\ell, \ell_0)$, where $\ell \sim (D/F)^\gamma$ is the characteristic distance between neighboring islands and $\ell_0 = (D/F)^{1/(2+d)}$ is the only combination of F and D with length dimension. If the system is smaller than ℓ_0 , it can accommodate at most one island, so that the island density cannot be obtained [17]. Recently it was shown [18] that there is another characteristic length in the system, called the layer-coherence length $\tilde{\ell} = \ell^{4/(4-d)}$, which is significantly larger than $\max(\ell, \ell_0)$. Systems smaller than $\tilde{\ell}$ exhibit perfect layer-by-layer growth, whereas larger surfaces show kinetic roughening. In our computer simulations we chose the system size larger than $\tilde{\ell}$ to prevent these finite size effects. Therefore it is essential to have an efficient simulation model.

Recently a very efficient coarse-grained Monte Carlo (CGMC) simulation method has been introduced [19]. The basic idea of the CGMC simulation method is that in order to measure the island density one needs not be able to follow the adatom diffusion on the atomic scale. Instead, one monitors the position of adatoms only on a much coarser scale, Δx , which must be smaller than the average distance between islands. Then, the computation time is reduced by about a factor $(\Delta x)^{-2}$.

We consider $L \times L$ cells on a two dimensional substrate, each of which is composed of $\Delta x \times \Delta x$ sites. For $i^* = 1, 2$ we used $L = 400$ and $\Delta x = 2$, whereas for $i^* = 3, 4$ the values were 200 and 4. Deposition of an atom into any of the cells happens with frequency $\nu_F = F(L\Delta x)^2$. With frequency $\nu_D = D(\Delta x)^{-2}$ all adatoms in the system are allowed to move to any of their neighboring cells or to stay where they are, with equal probability. If there are no adatoms in the system, $\nu_D = 0$. In each Monte-Carlo step either an atom is deposited with probability $\nu_F/(\nu_F + \nu_D)$, or all adatoms are allowed to move. The motion of adatoms within the cells is ignored in the coarse grained picture. Instead, the state of each cell is characterized by three variables: First, $h(x)$ is the number of completed monolayers which corresponds to the height in cell x . Second, $0 \leq m(x) < (\Delta x)^2$ is the number of

atoms on top of the $h(x)$ monolayers. Finally we assign a step indicator “flag” to each cell, which is either 0 or 1, indicating whether the m atoms belonging to the cell are mobile or immobile, respectively. They are immobile, if the cell contains an island edge, because for simplicity we assume that then the adatoms are irreversibly bound there, before they move out of the cell again. An island edge can be created within a cell, if $i^* + 1$ atoms are gathered there, so that a stable island nucleates. It may also happen that an island fills a whole cell and starts invading the neighboring cells, whose step indicator is then set equal to 1 without nucleation.

In a deposition step, a cell is chosen at random, and its number m of atoms incremented by one. If m then exceeds i^* , a nucleation event is recorded, and the flag is set equal 1. The diffusion process can be implemented in two different ways. In previous work all adatoms were simultaneously allowed to move into neighboring cells (parallel updating of the list of mobile atoms). Afterwards it was checked, which of the cells contained more than i^* atoms, and the corresponding nucleation events were recorded.

Here, we implement the diffusion process in a different way. As before the mobile atoms are elements of a list. However, now starting with the first element the atoms are allowed to diffuse to a neighboring cell sequentially: After each move it is checked whether it results in a nucleation event, i.e. whether m exceeds i^* . In this case all m atoms form a stable island and are deleted from the list of mobile atoms. The holes in the list are filled with the last elements, which leads to a randomization of the list. Then the next atom on the list is allowed to move, it is checked, whether this triggers a nucleation, and the list of mobile atoms is updated accordingly. In this way we continue until we went through the whole list once.

Fig.1 illustrates the difference between the parallel and the random sequential way of updating the list of mobile atoms for the case $i^* = 1$: For parallel updating it may happen that two atoms just exchange their positions. Instead, in reality they are so close to each other that they could easily meet to form an island. The random sequential update takes this into account.

In our simulations we recorded the number of islands at a coverage 0.12. The two different ways of updating in the diffusion process give different values for the exponent γ . It turns out that the random sequential update produces values, which are closer to the theoretical ones than those obtained by the parallel updating [17] and other methods [20], especially for larger stable island sizes. The numerical values we obtained are compared with other values in the following table.

In the table, the values in the column (1) are from Eq. (1) with $d_f = 2$. The values in the column (2) are from Eq. (1) with $d_f = 1.7$. The values in the column (3) are the ones from Ref. 8 using the parallel updating. The values in the column (4) are the one obtained by Family and Amar [20] using the kinetic Monte Carlo method [21]. The values in the column (5) are the ones we obtained using the random sequential updating and a linear fit to the double-logarithmic plot of the data (see Fig. 2) excluding the ones for $D/F \leq 10^6$. Column (6) contains a fit of the same data points with the formula

$$\rho = \text{const}(D/F)^{-2\gamma}(\log D/F)^{-1/3}, \quad (2)$$

which shows that the logarithmic correction worked out by Tang [14] does not change our conclusion that the random sequential update gives better agreement with the theoretical values for the exponent.

In summary, we have investigated the island density in the submonolayer regime of homoepitaxial growth. The island density on a two dimensional substrate decreases with increasing the ratio D/F as $\rho \sim (D/F)^{-2\gamma}$. The exponent γ depends on the critical size i^* . Performing the coarse grained Monte Carlo simulation with random sequential updating in the diffusion step, we have obtained numerical values of the exponent $\gamma(i^*)$ for $i^*=1, 2, 3$, and 4 which are in better agreement with the theoretical values than the ones obtained previously by other methods.

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TABLES

i^*	(1)	(2)	(3)	(4)	(5)	(6)
1	0.333	0.35	0.344	0.33	0.336	0.336
2	0.500	0.52	0.48	0.5	0.507	0.523
3	0.600	0.62	0.55	0.58	0.609	0.592
4	0.667	0.68	0.58		0.658	0.650

TABLE I. The comparison of the numerical values of 2γ with the theoretical values and the ones obtained by other simulation methods. The values in the column (1) are from the theoretical formular Eq. (1) with $d_f = 2$. The values in the column (2) are from Eq.(1) with $d_f = 1.7$. The values in the column (3) are quoted from Ref.8 using the parallel updating. The values in the column (4) are quoted from Ref.20 using the kinetic Monte Carlo method. The values in the column (5) are the ones we obtained using the random sequential updating. The values in the column (6) are the ones fitted by the formular (2) with the logarithmic correction.

FIGURES

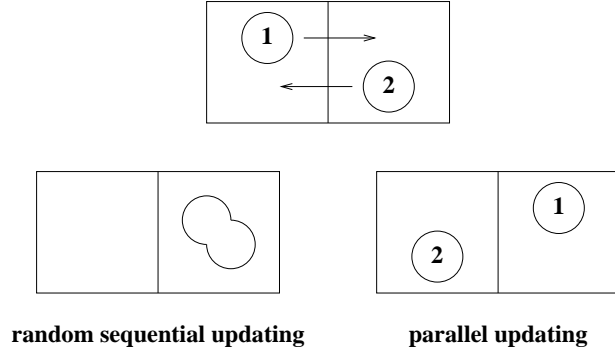


FIG. 1. Difference between the random sequential and the parallel updating rule. In the random sequential updating, particle 1 moves first and meets particle 2, and nucleation occurs ($i^* = 1$). But in the parallel updating, particles can collide but cross each other without nucleation.

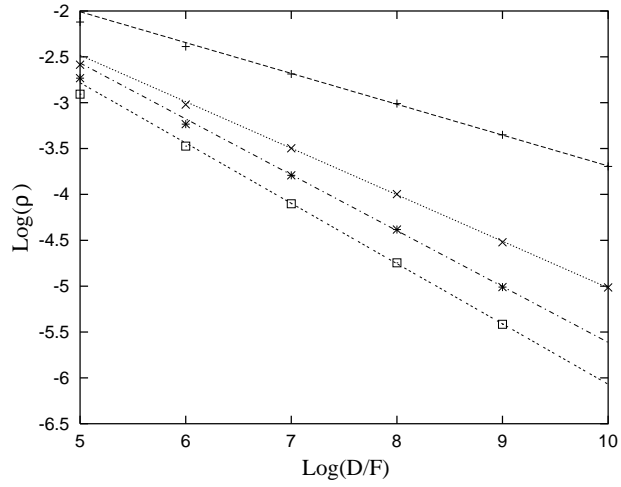


FIG. 2. The density of islands versus the ratio D/F in log-log scale for the cases of $i^* = 1, 2, 3$, and 4 from the top.